# Coupled THMC Models for Bentonite in Clay Repository for Nuclear Waste: Illitization and Its Effect on Stress under High Temperature

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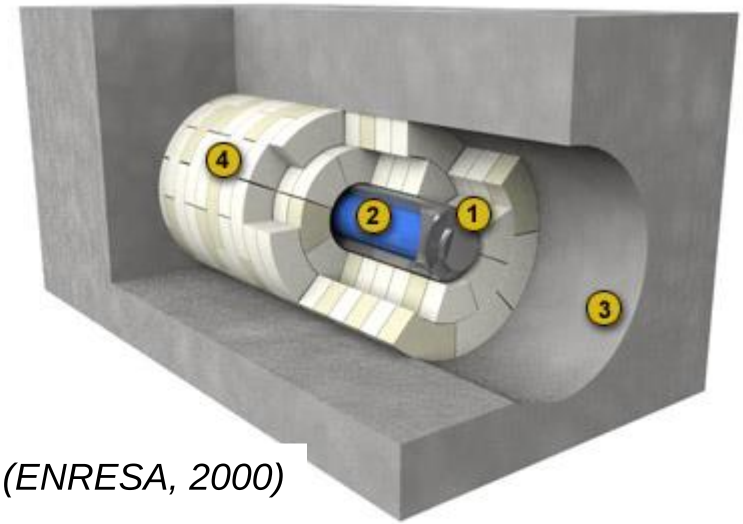
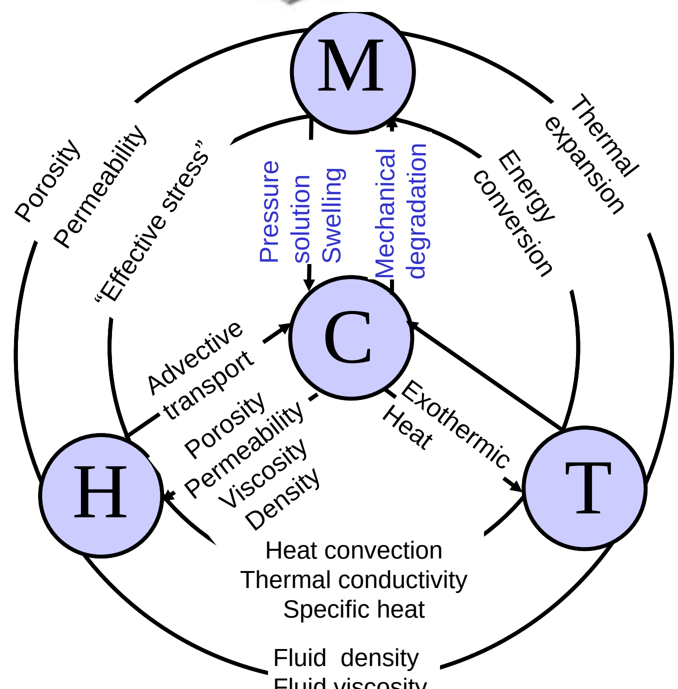
# Background (1)

- Engineered Barrier System (EBS) which is typically composed of bentonite is one important component in nuclear waste disposal
- Clay rock is one important type of hosting formation *for nuclear waste disposal* such as Opalinus Clay at the Mont Terri site, Switzerland.
- Swelling in the bentonite EBS and the clay rock are important as it could enhance the stability of the emplacing drift and seal fractures in the host rock
- However, the swelling capacity is reduced by illitization, the transformation of smectite to illite.
- Illitization is affected by the availability of K, Al, water and temperature
- THMC processes are tightly coupled in bentonite and clay formations. To study the illitization in the bentonite and clay rock and its effect of the swelling requires a coupled THMC model




*(ENRESA, 2000)*



# Background (2)

## Why do we study coupled THMC processes at high temperatures (>100 $^oC$)?

- A thermal limit of about 100°C is currently imposed in all disposal concepts throughout the world that involve the use of bentonite buffer and backfill materials. Chemical alteration and the subsequent changes in mechanical properties are among the determining factors.
- However, the thermal limit of 100°C might be overly conservative
- The impact of higher temperature on bentonite behavior and the consequences on repository performance, remain largely open questions.
- Thermal limit is of particular importance for clay reposition because of the low thermal conductivity of clay formation.

# **Model Development**: Simulator

*TOUGHREACT-FLAC3D which combines TOUGHREACT and TOUGH2-FLAC3D has been developed and it is capable of conducting coupled THMC simulations.*

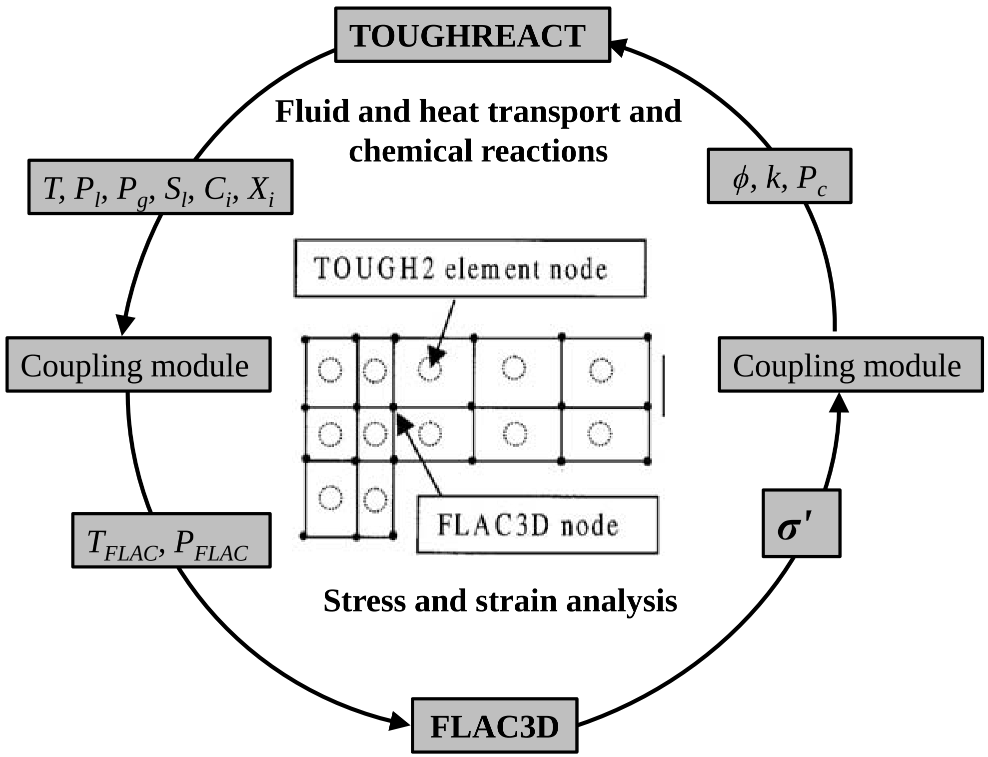
***TOUGHREACT — coupled THC code***

*TOUGHREACT is a numerical simulator for chemically reactive nonisothermal flows of multiphase fluids in porous and fractured media(Xu et al., 2011)*

***TOUGH2-FLAC3D — coupled THM***

TOUGH2-FLAC3D *(Rutqvist, et al., 2011) sequentially couples the finite-difference geomechanical code FLAC3D with the finite-volume, multiphase fluid flow code, TOUGH2 (Pruess et al., 1999), it* has been successfully used in different applications including geothermal energy development and CO2 geological sequestration.

*THC Processes*




*Mechanical Process*

# ***Model Development:*** *MC coupling*

An extended linear elastic swelling model for EBS bentonite

$$d\sigma_s = \boxed{3K\beta_{sw} ds_l} + \boxed{A_n dC} + \boxed{A_{sc} dm_s}$$

*Stress change due to water saturation change*

*Stress change due to chemical concentration change*

*Stress change due to smectite abundance change*

M C coupling through link chemistry to mechanics through the micro-structure strain based dual structural Barcelona Expansive Clay Model

$$\mathrm{d}\varepsilon_{vm}^{e} = \frac{f_s}{K_m}\mathrm{d}\widehat{p} \qquad K_m = \frac{e^{-\alpha_m \hat{p}}}{\beta_m} \qquad \widehat{p} = p + s_m \qquad s_m = s + s_o$$

*The effect of ionic strength is accounted through osmotic suction*

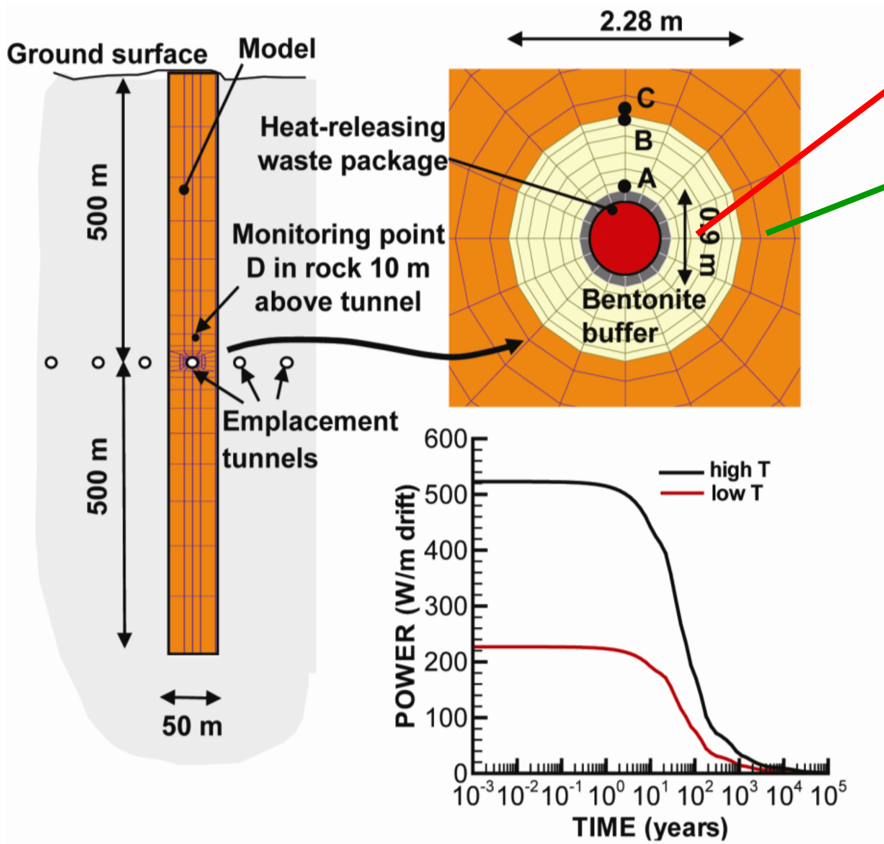
$$s_o = -10^{-6}\frac{RT}{V_w}\ln(a_w)$$

*The effect of exchangeable cations is accounted through*

$$\beta_m = \sum_i \beta_m^i x_i$$

*The effect of the amount of smectite is accounted through the mass fraction of smectite* $f_s$

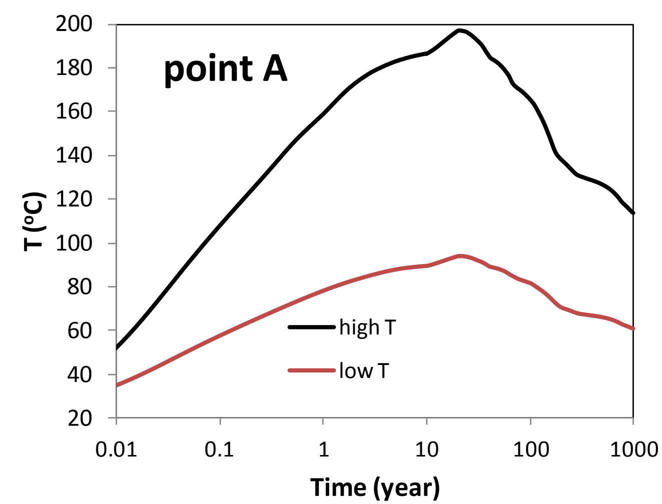
# Model Development:
## Modeling Scenarios




***EBS Bentonite: Kunigel-V1 and FEBEX bentonite***

***Clay formation : Opalinus Clay***

*Two cases for comparison: a "high T" case and a "'low T" case*

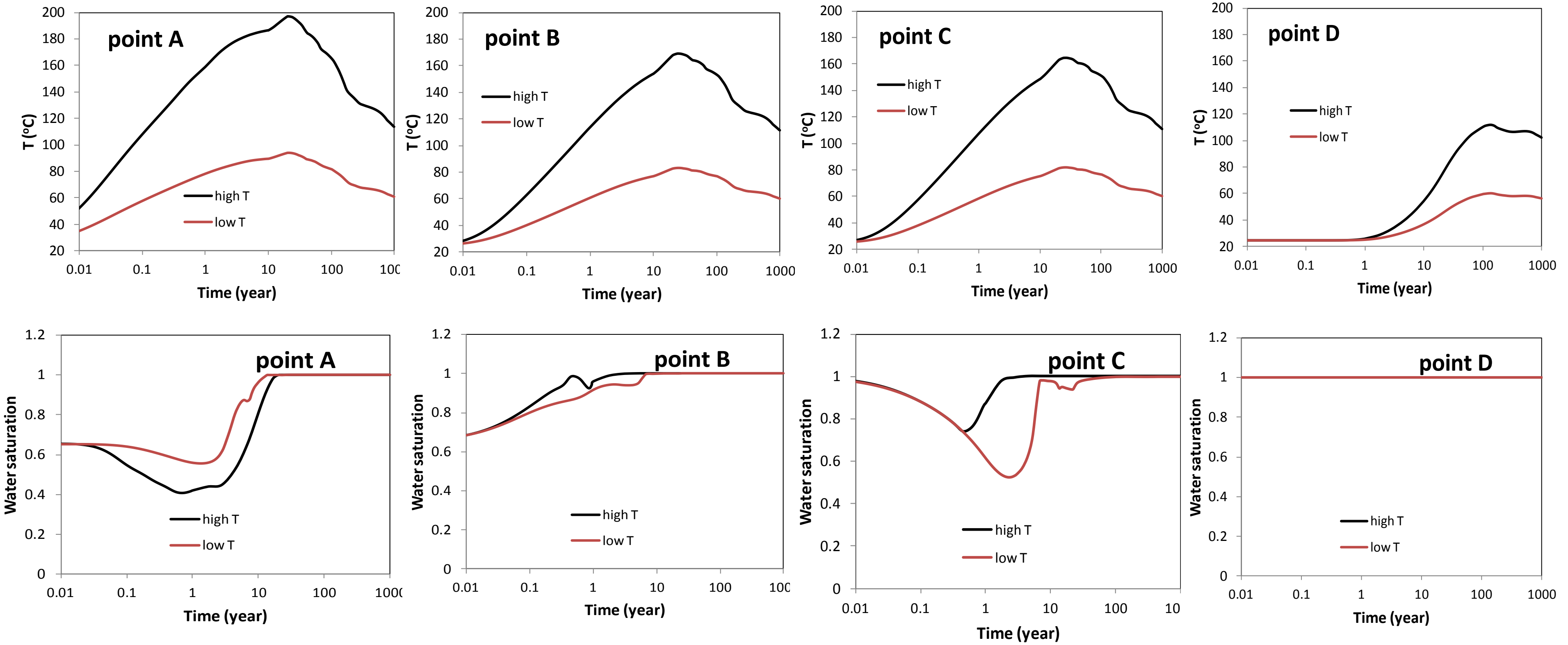


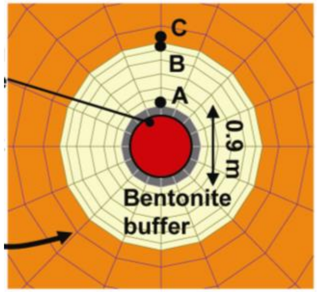

- *Chemical model: 12 primary species, 97 aqueous complexes, 17 minerals and 5 exchangeable cations*
- *Illitization can be modeled as smectite dissolution and neo-formation of illite:*

*Smectite + $0.52H^+$ + $0.63AlO_2^-$ + 0.6K = illite + $0.26H_2O$ + $0.08Mg^{+2}$ + $0.33Na^+$ + $0.5SiO_2(aq)$*

- *The reaction rate from 4.5e-14 to 2.4e-13 mol/g/s calibrated against data from Kinnekulle bentonite, Sweden (Push&Madsen, 1995)*

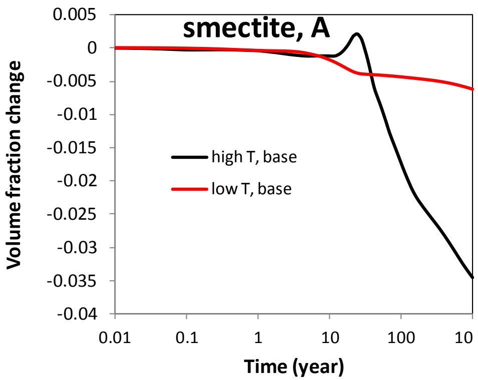
# Model Results:
## Temperature and Water Saturation Evolution

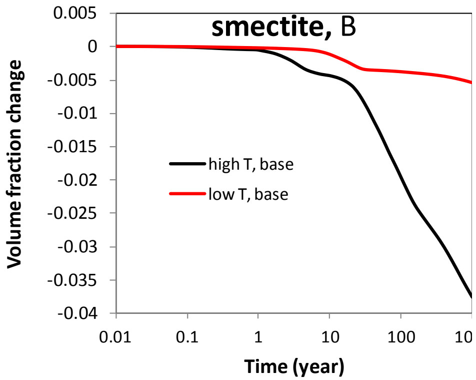


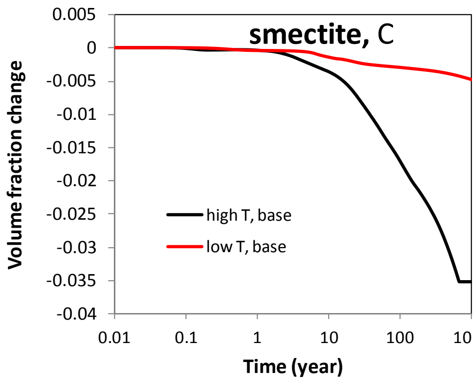



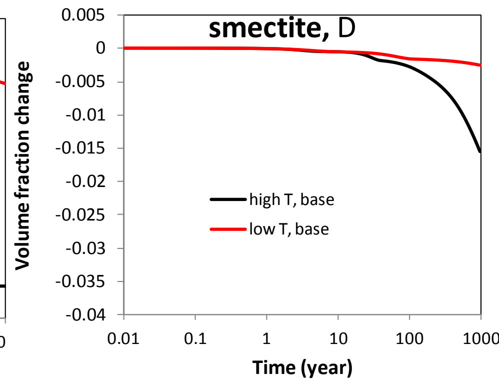

*Temperature and water saturation evolution at points A, B, C, and D.*

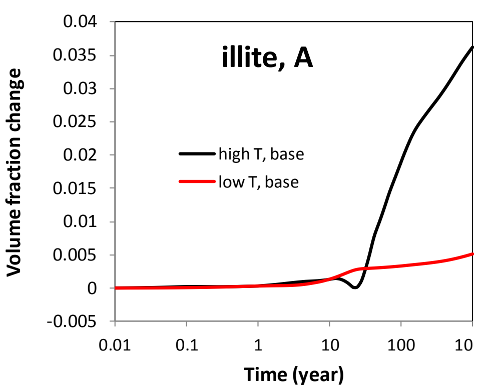
# Model Results for Kunigel-VI Bentonite : Illitization

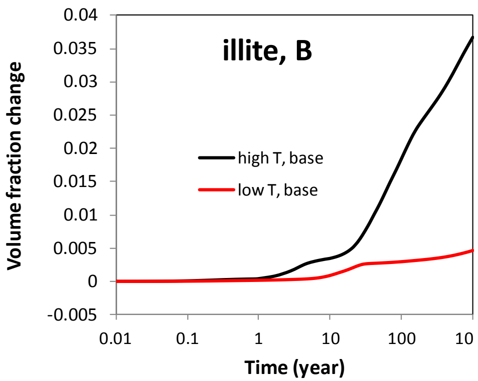


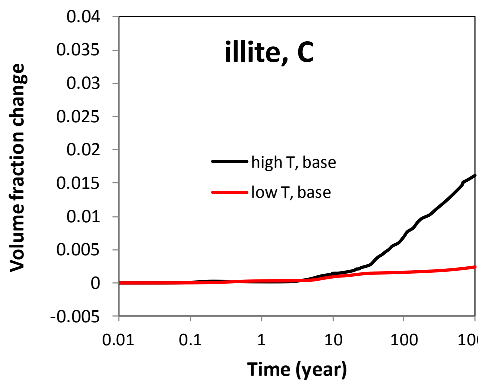



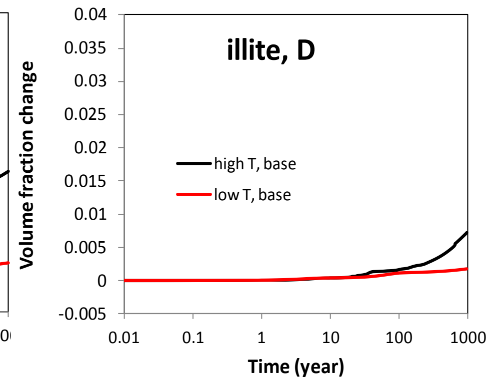



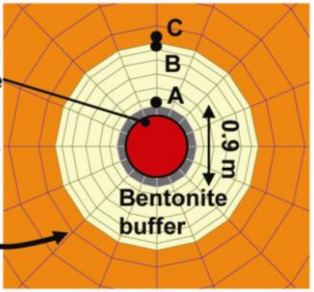



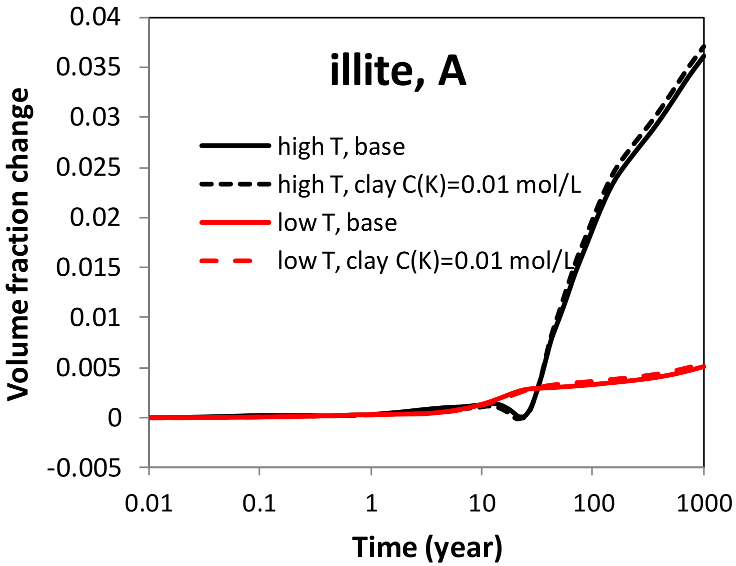



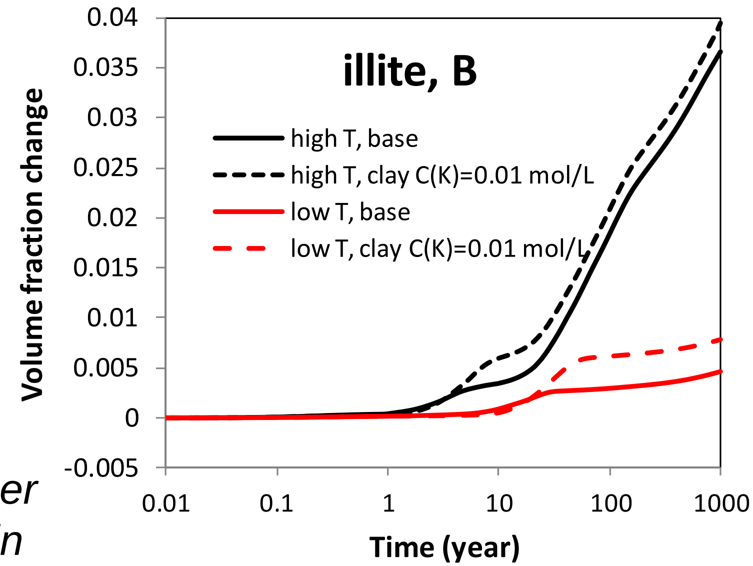




*Smectite volume fraction in EBS bentonite decreases by 0.035, or 11% of the initial amount (0.314) for "high T" case (VS 1.5% for "low T" case) after 1000 years*

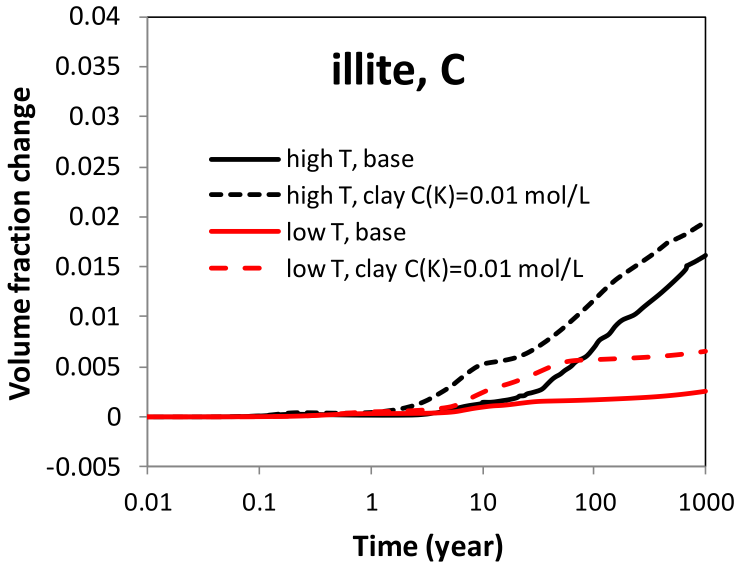
*In clay formation, a 100% loss of smectite near the bentonite-clay formation interface and 43% loss 10 m away from the interface after 1000 years*

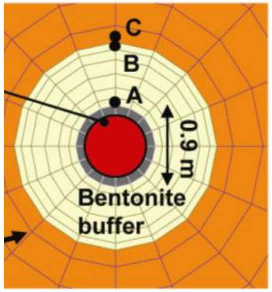
# Model Results for Kunigel-VI Bentonite:
## The Effect of Aqueous Composition

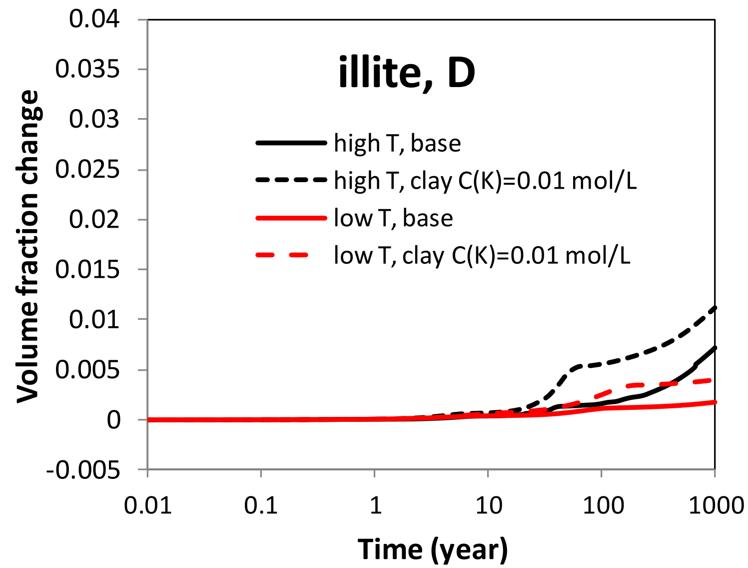


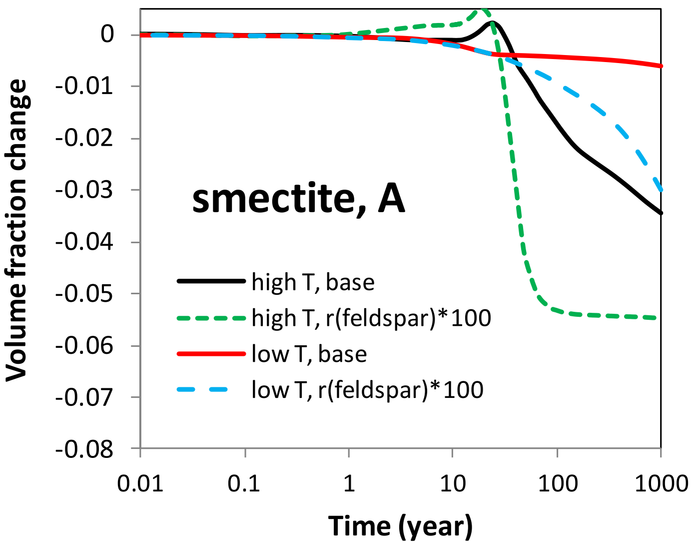

*Models with higher K concentration in the clay formation*



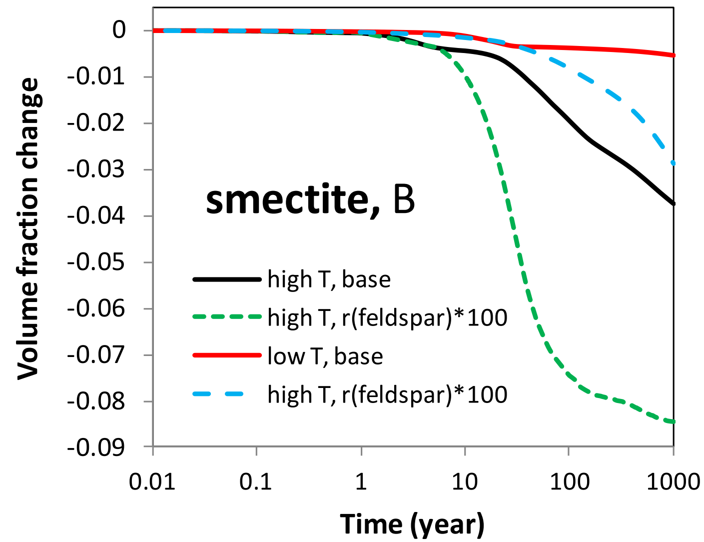



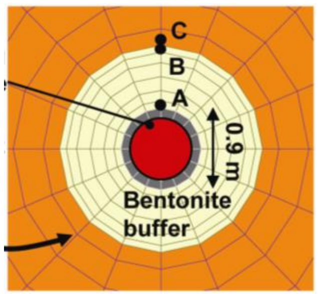
# Model Results for Kunigel-VI Bentonite: The Effect of Accessory Minerals

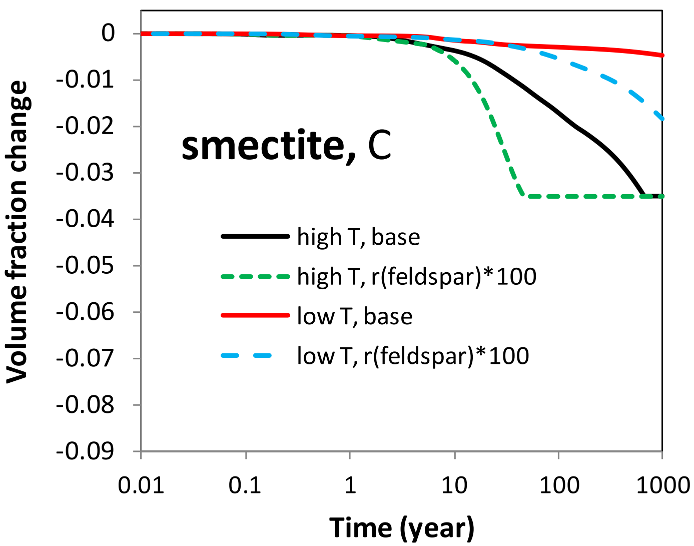



*K-feldspar = $AlO_2^-$ + $K^+$ + $3SiO_2(aq)$*

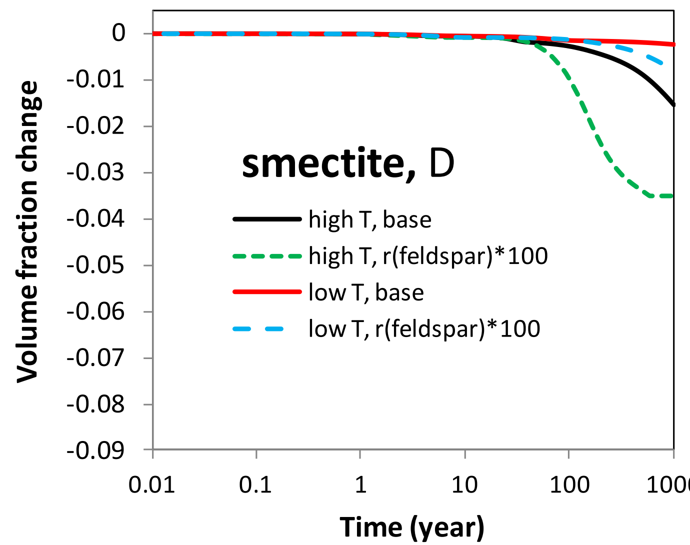
*Model with K-feldspar dissolution rate two orders of magnitude higher*



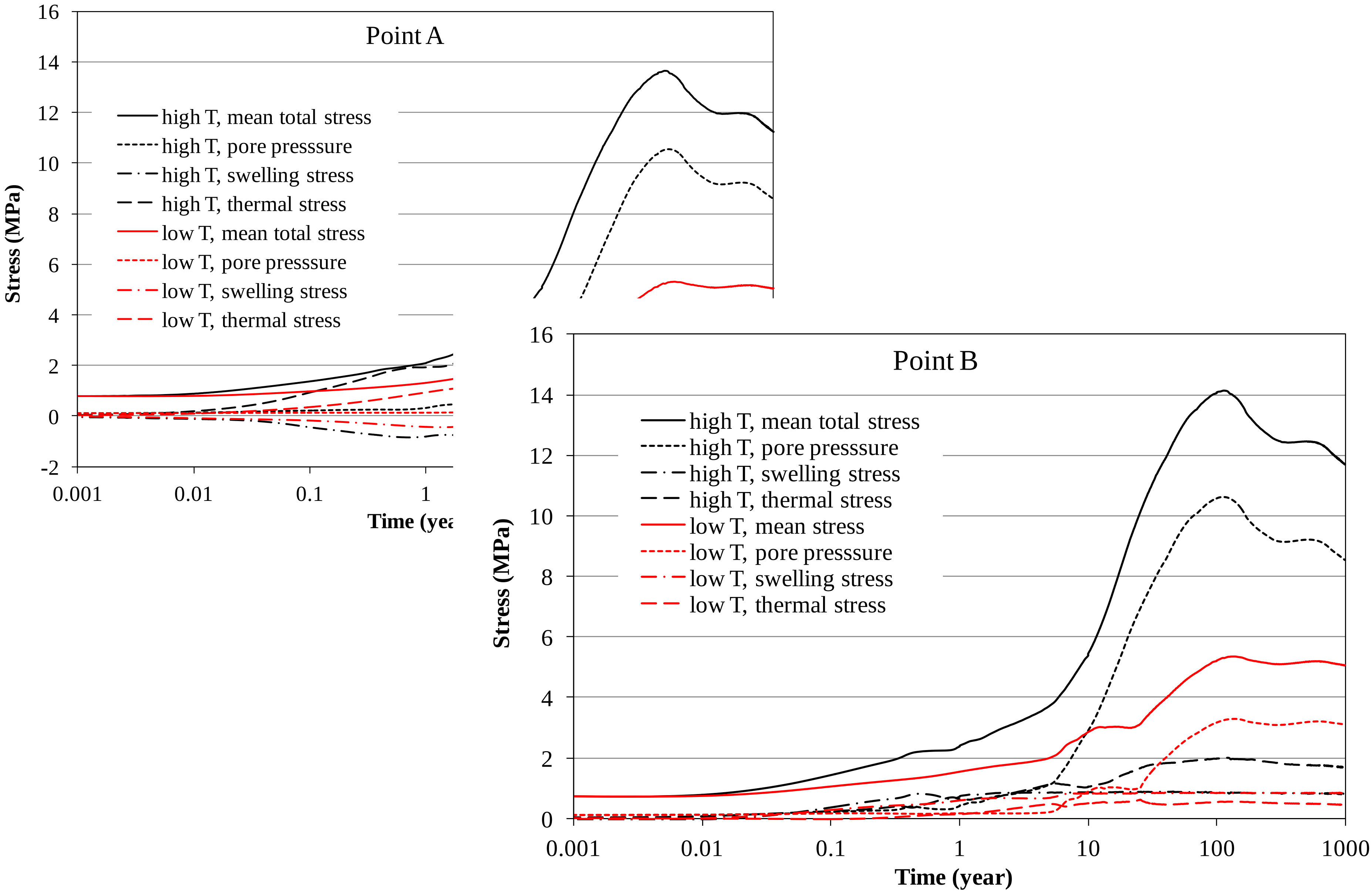



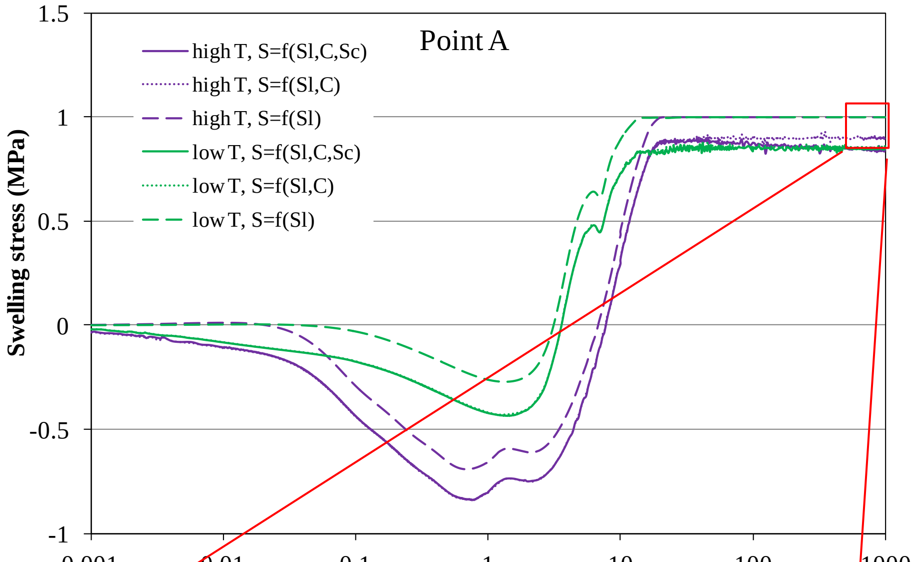

*18-27% smectite dissolves when K-feldspar dissolution rate is higher, VS 11% in the base case*

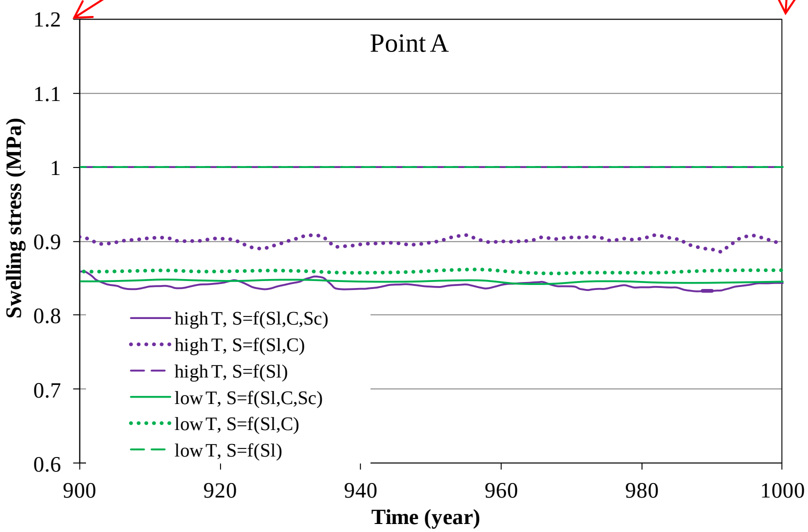


# Model Results for Kunigel-VI Bentonite: Stress Analysis



# Model Results for Kunigel-VI Bentonite:
## Chemical Effect on Swelling Stress




$$d\sigma_s = 3K\beta_{sw} ds_l + A_n dC + A_{sc} dm_s$$

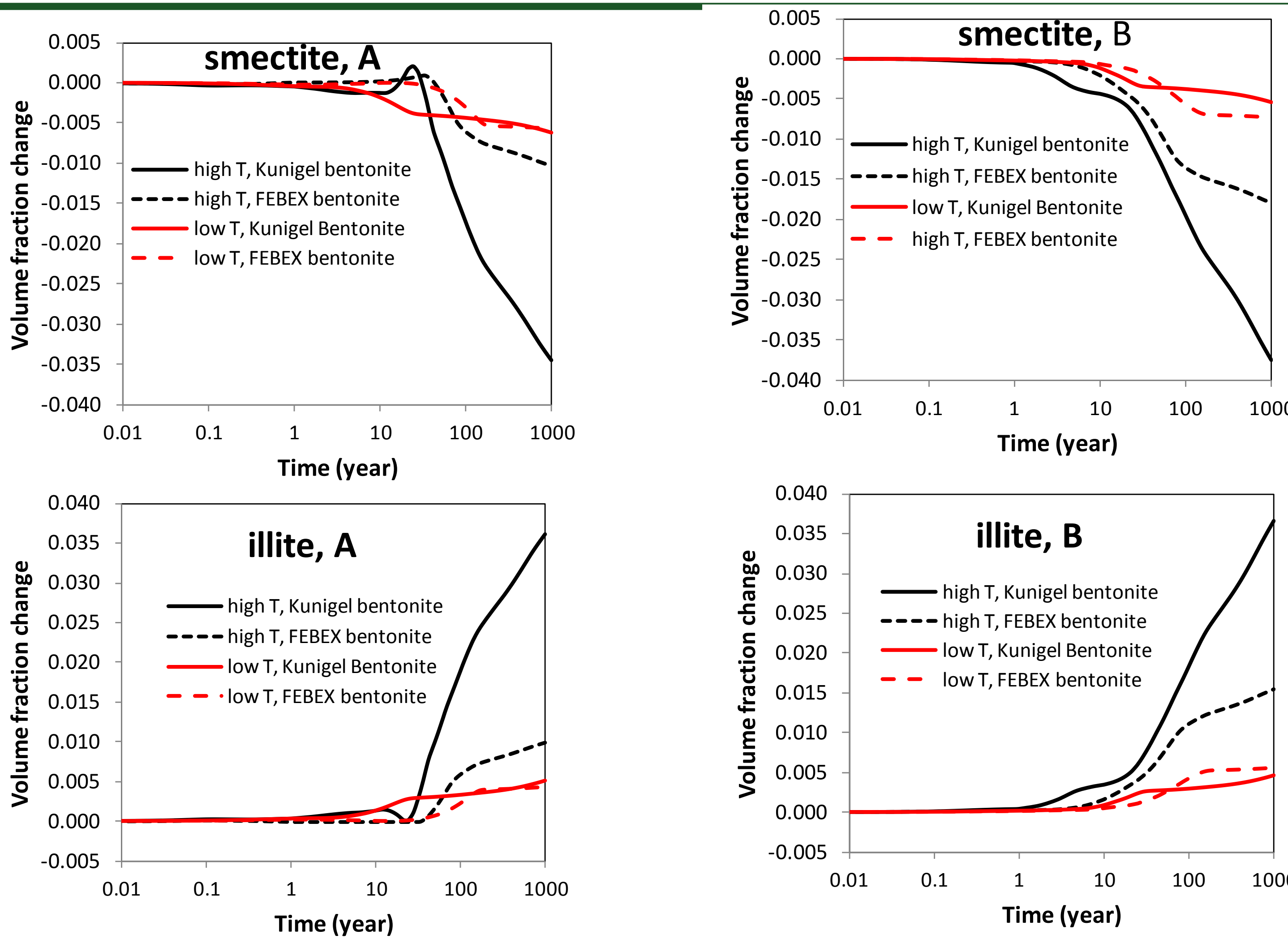
- *"S=f(Sl,C,Sc)", the swelling stress is calculated as a function of liquid saturation changes (Sl), ion concentration (C) changes, and smectite (Sc) changes.*
- *"S=f(Sl,C)", the swelling stress is only a function of liquid saturation and ion concentration In the third set, denoted as "S=f(Sl)",the swelling stress is only a function of liquid saturation changes.*

*"high T" base case: 16-18% reduction in swelling stress*

*"high T" "r(feldspar)*100" case: 20-26% reduction in swelling stress*

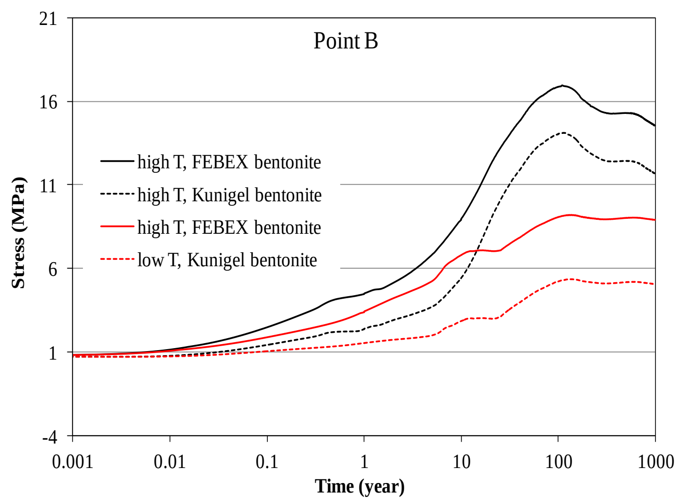
# Model Results for FEBEX bentonite:

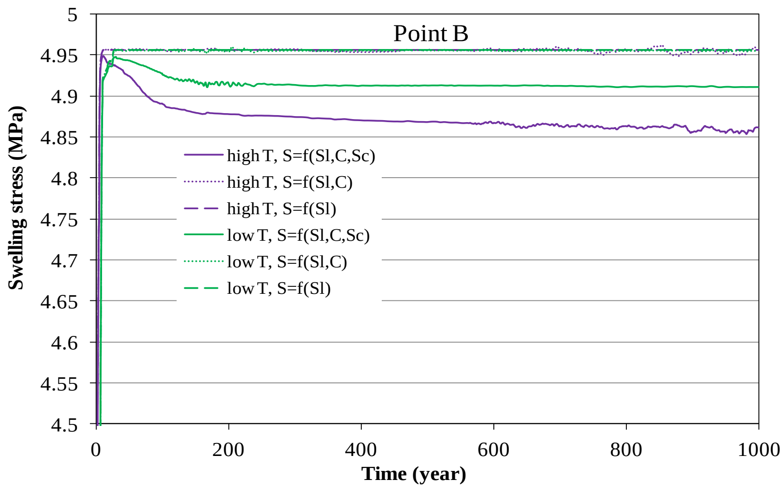
## Illitization




*FEBEX bentonite undergoes less illitization than Kunigel-VI bentonite due to high concentration of cations in pore water and less K-feldspar in solid phase.*

# Model Results for FEBEX Bentonite:
## Stress Analysis




| Kunigel bentonite | | | | FEBEX bentonite | | | |
|---|---|---|---|---|---|---|---|
| Stress reduction by ion concentration, MPa | Stress reduction by ion concentration, (%) | Stress reduction by smectite dissolution MPa | Stress reduction by smectite dissolution (%) | Stress reduction by ion concentration, MPa | Stress reduction by ion concentration, (%) | Stress reduction by smectite dissolution, MPa | Stress reduction by smectite dissolution (%) |
| 0.1 | 10% | 0.05 | 5% | 0.006 | 0.1% | 0.076 | 1.5% |

# Summary

- *Coupled THMC models were developed to evaluate the chemical alteration and associated mechanical changes in a generic repository.*
- *In general, illitization in the bentonite and clay formation is enhanced at a higher temperature. However, the quantity of illitization is affected by many chemical factors and subsequently varies a great deal, important chemical factors are the concentration of K, Al and dissolution rate of K-feldspar.*
- *In 1000 years for the 200°C scenario, smectite in Kunigel-VI bentonite decrease 0.4 – 8.5 vol% (about 27% of the initial amount) whereas smectite in FEBEX bentonite decrease 1 – 4 vol% (7% of the initial amount).*
- *Higher temperatures lead to much higher stresses in the near field, caused by thermal pressurization and vapor pressure buildup in the EBS bentonite and clay host rock.*
- *Chemical changes including changes in pore water ion concentration and smectite volume fraction, however, lead to a reduction in swelling stress, which is more pronounced for Kunigel-VI bentonite than for FEBEX bentonite: . 16–18% reduction for Kunigel-VI bentonite versus 1.5–3.6% for FEBEX bentonite  for the 200°C scenario.*

# Ongoing and Future Work

- *Considering illitization through solid state transformation by substitution of intracrystal cations*
- *Developing more rigorous approach to link chemistry to mechanics for more accurate calculation of the mechanical-chemical coupling in bentonite*
- *Taking into account of chemical changes in the canister. In the current model, the canister serves only as a heat source. Further model analysis is needed to consider chemical changes in the canister, specifically the release of $Fe^{+2}$, which might enhance the dissolution of smectite by forming chlorite*

## *ACKNOWLEDGMENTS*

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# Thanks for your attention!
# Questions?